Drinovec et al.: Traceable calibration of in-situ aerosol absorption instruments with monodisperse nigrosin, egusphere-2026-1501

Review

General

The paper presents analyses of calibration methods of aerosol light absorption measurement instruments applied to a 2-wavelength photo-thermal aerosol absorption monitor PTAAM. The blue wavelength 450 nm was calibrated with a well-defined mixture of NO2 and synthetic air and by using published absorption spectra so that is basically like the calibration of any gas monitor, there is nothing to comment.

The more interesting and important part of the paper is the calibration of the near-infrared wavelength 808 nm. Nigrosin was nebulized and lead selectively either through a DMA, a CPMA or their combination in a tandem arrangement either to an SMPS, the PTAAM, a CPC or an FCAE. With these instruments size dependent absorption coefficient was obtained both as measured with the PTAAM and modeled using a Mie code. In addition, the size-dependent density of the nigrosin particles was determined. Different instrument combinations for calibrations were tested and the uncertainties were analyzed.

This is an excellent and useful paper for anyone building a calibration setup, perfectly in line with the objectives of the funding STANBC project. I don't find any major errors in it but I do find something to modify. As it will very likely be used as a reference for other groups, both experiencd and those new in the field building their calibration setup, I wish to see some more more details that would help building and repeating the measurements and also understanding them. For example, to be able to repeat the measurements reported here it would be very useful and important to see a more detailed schematic than in the present Figs. 2, 5, 7 and 9. I suggest you draw a bit more detailed figure showing tube diameters, flows, valves etc. that you used, either in the paper or in the supplement. I was also missing scatterplots of Mie-modeled absorption vs. measured absorption with slopes, offsets, r^2 and p-values to all the figures where you now have the bar charts (Figs. 6, 8, 10, S6). They would be all very informative.

I will present questions, suggestions and wishes in the detailed comments below, I leave it to the authors to decide whether they want to revise the paper accordingly.

Detailed comments

L141-146: It is first written "Typical instrument noise under controlled laboratory conditions (using filtered room air) is shown in Fig. 1 with 15 min average noise of 0.03 Mm^{-1}." Then it is written "Actual measurement uncertainty for 15 min experiments of 0.3 Mm^{-1} was determined by the variation of the absorption coefficient for the filtered sample". Quite a big difference. You measured filtered air in the lab before and during the actual calibration experiments but the noise was an order of magnitude higher in the latter. Was everything – especially the flow through the PTAAM really the same except the connections to the other instruments in the calibrations? I Would it make sense to plot two more lines in Fig. 1, those valid for the actual measurements?

Fig 1. Define tau in the caption.

L 155-158 I think it would be useful to explain and emphasize here that the CPMA classifies particles according to their mass-to-charge ratio which is essential especially for the experiments presented in this paper. In the present version of thé paper I don't find the words " mass-to-charge ratio " at all.

L192 "Because the absorption coefficient depends mainly on particle mass " That is not quite correct. There is size dependence, see for instance Fig. 4 in Bond and Bergstrom, Aerosol Science and Technology, 40:1, 27-67 (2006) and the explanations on the same page.

L192-193 "... we decided to use the mass equivalent particle diameter D_m ..." Please write the equation. The point is that for the equivalent diameters several symbols and equations have been used in the literature which is a bit confusing. I collected some at the end of this review. So, it would be most useful for future readers if you wrote this as an equation.

Table 1, caption "Measured mobility diameter D_p, effective density and mass-equivalent diameter D_m" See DeCarlo et al. (2004), eq.(25) they use d_m for electric mobility diameter, their mass equivalent diameter is d_{me}, eq.(1), and d_p is their physical diameter, p 1186, also in textbooks. See again the list I collected at the end of the review.

L213-214 "...corrected for particles corrected for multiply-charged particles and diffusion losses. " Did you use penetration efficency data in Fig. S7 or were the losses corrected using equations published in the literature? If published equations were used, please give the citations.

Fig 4. A suggestion: how about showing also absorption size distribution.

L253-255 "The absorption was then calculated as the sum of the absorption of singly-charged particles with mass m_0, doubly-charged particles with mass $3.5*m_0$ and triply-charged particles with mass $6.6*m_0$." I don't understand. Explain, how you got the multipliers 3.5 and 6.6 detailed enough for others to repeat.

L255-256 " multiply-charged particles accounted for 15% and 25% of the total light absorption." Are these numbers for doubly and triply charged particles or what? How did you get them? Another thing is that I suppose you didn't make just one scan so these numbers are probably averages of some number of measurements. Am I right? Then give ave +- std or some other uncertainty measure. I find it always a bit suspicious if people give some integer numbers as the only result of any measurement.

L257-259 "... relatively broad mass distribution (Rm=mp/FWHM=1.5) is used. This was achieved by sample-to-sheet flow ratio of 1.5 L min^{-1}/5 L min^{-1}...." I thought you simple select Rm at the user interface of the CPMA. And reading the CPMA-related literature setting Rm is not as simple as setting the sample-to-sheath ratio. Am I wrong? Please cite the ref where this is given. And correct sheet to sheath.

Fig. 6. Why don't you plot also Mie(D_p)?

L270-273 I think it would be useful especially for non-CPMA users to remind here that the CPMA selects particles according to their mass-to-charge ratio.

L307-308 " For example, we set the CPMA to select 1 fg particles, then the DMA should be set to mobility diameter of 110.4 nm for nigrosin particles." Explain the calculation so that readers can replicate it.

L312-313 "... for m_0=1 fg we obtain a mobility diameter of 93 nm and 80 nm for doubly- and triply- charged particles." Explain the calculation so that readers can replicate it.

L336-338 "The combined uncertainty is calculated as the square-root of the linear sum of squared standard uncertainty components for a coverage factor k=1 (68 % confidence interval)." Show also as an equation. Explain coverage factor, I have never encountered it and I guess I am not the only one.

L372-373 "To determine the correct particle absorption coefficients, a particle mass (or mass-equivalent diameter) should be used in the Mie calculations." I would be a bit hesitant to make such a strong statement. Interactions of light and particles are determined by the physical size after all. Yes, the modeled absorption agreed very well with the measured when the mass-equivalent diameter was used for the modeling. But still I am wondering whether there could be an explanation for the higher absorption coefficients when you used the electric mobility diameter for the modeling. At least try to give some possible explanations

SUPPLEMENT

Figure S3. Explain the vertical lines. I have also a suggestion: you could actually add vertical lines for 1 fg and 2 fg and the corresponding horizontal lines and also write the MAC values to explain the text on lines 275 – 276 of the main paper.

L73 "NO_2 loses" > NO_2 losses

L78-90 The penetration efficiency was calculated from number concentration. For the purpose of the present paper I think it could be useful to calculate it from also from size-dependent absorption coefficient. It could in theory even explain why the Mie modeled absorption is higher than the measured. Think about it.

Fig. S7 Why is the efficiency so noisy for D_p > 300 nm? Does it have any effects on the data analyses of the paper?

Some symbols and names used for particle equivalent diameters in the literature

d_p = physical or geometric diameter, Baron and Willeke (2001); DeCarlo et al. (2004); Baron et al. (2011)

d_m = mobility diameter = $((6/\pi)(m/\rho_{eff}))^{1/3}$ from Eq. (2) of Johnson et al. (2013)
d_m = electrical mobility diameter, DeCarlo et al. (2004), p. 1190, Eqs. (24) and (25)
d_m = mass equivalent diameter = $((6/\pi)(m_p/\rho_m))^{1/3}$, from Eq. (23-1) of Baron et al. (2011)

d_{me} = mass equivalent diameter = $((6/\pi)(m_p/\rho_{eff}))^{1/3}$ from Eq. (3) of DeCarlo et al. (2004)
d_{me} = electrical mobility equivalent diameter, Olfert and Collings (2005)

d_B = electric mobility diameter, Baron et al. (2011) p. 513, Eqs. (23-12) and (23-13)

References

Baron and Willeke (eds.), Aerosol measurement: principles, techniques, and applications, 2nd ed., Wiley, 2001.

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DeCarlo et al., Particle Morphology and Density Characterization by Combined Mobility and Aerodynamic Diameter Measurements. Part 1: Theory, Aerosol Science and Technology, 38:1185–1205, 2004

Johnson et al., Mass–Mobility Measurements Using a Centrifugal Particle Mass Analyzer and Differential Mobility Spectrometer, Aerosol Science and Technology, 47:1215–1225, 2013.

Olfert and Collins, New method for particle mass classification — the Couette centrifugal particle mass analyzer, J. Aerosol Science 36, 1338–1352, 2005.